Exchange Split Quantum Well States of a Noble Metal Film on a Magnetic Substrate

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By spin resolved photoemission we have detected exchange-split spin-polarized electronic states in a nonmagnetic material deposited on a magnetic substrate. Epitaxial Cu overlayers on fcc Co(100) exhibit *sp* symmetry electronic states which are spin polarized by the confining magnetic interface. These spin-polarized states can be observed up to a film thickness of at least 10 atomic layers. The spectral polarization and intensity at the Fermi level display a periodicity correlated with the oscillations of the long range coupling between magnetic films separated by Cu(100) layers.

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Ultrathin layered systems enable the synthesis on an atomic scale of new and complex magnetic structures. New perspectives in this field have been opened by the discovery of the long range exchange coupling between magnetic films separated by a nonferromagnetic layer [1,2]. The sign of the magnetic coupling exhibits an oscillatory behavior for varying thicknesses of the nonmagnetic spacer [3]. The oscillatory and long range character of the coupling suggests that its origin can be understood within the frame of RKKY interactions. Theoretical analysis based on the RKKY coupling scheme relate the period of the oscillations to the dimensions and topology of the Fermi surface of the nonmagnetic metal [4,5].

Recent photoemission and inverse photoemission experiments [6,7] from noble metal overlayers on ferromagnetic materials have been interpreted in support of this RKKY coupling mechanism. Some *sp*-derived electronic states of the noble metals show a simple periodic pattern as a function of the layer thickness, which appears to be correlated with the oscillatory behavior of the magnetic coupling. Moreover, these states present some of the characteristic features of one-dimensional quantum well states and their wave vector can be related within a simple picture to the wave vectors of the RKKY coupling.

This unusual set of properties suggest thus that the electronic states responsible for the magnetic coupling could become accessible to spectroscopic investigation. The key question which still needs to be addressed is whether the "quantum well states" of noble metal overlayers are spin polarized. Spin-polarized states could obviously mediate the coupling between the magnetic layers. A conclusive evidence for spin polarization has not been provided by the experiment of Ortega and coworkers [6,7] which was performed without spin analysis. An indication for the magnetic character of these quantum well states is given by a spin-down state reported for low Ag coverage on Fe(100) [8]. According to Ortega and co-workers [6,7] this level could eventually develop into a polarized quantum well state. However, this state, which is identified as a Fe 3*d*-derived interface state on the basis of a tight-binding calculation, is experimentally observed only up to 3 Ag monolayers [8].

Remarkably, no oscillations of the spin polarization associated with the long period of the exchange coupling through Ru could be detected in a recent experiment by spin-polarized Auger spectroscopy [9]. The Auger polarization of a Ru overlayer on Fe(100) shows that the induced spin polarization on the nonmagnetic material is concentrated at the interface. Away from the interface the Ru polarization decays rapidly. It can be described either by a steep decrease or by strongly damped short period oscillations. These results thus raise some questions on the relation between polarization and exchange coupling across a nonmagnetic layer.

The purpose of this paper is to report on the direct observation of spin-polarized states in a noble metal overlayer. We have investigated the electronic structure of Cu(100) overlayers on fcc Co(100) by spin- and angle-resolved photoemission with synchrotron radiation. In agreement with earlier spin-integrated photoemission experiments, some states display a periodicity correlated with the oscillations of the magnetic coupling. *These states of the Cu film are found to be spin polarized up to at least a 10 monolayer thickness.* Within a simple quantum well state picture and in the asymptotic high-coverage limit the periodicity of these polarized states corresponds to the dominant RKKY oscillation along the ⟨100⟩ direction. The data also demonstrate that discrete polarized states of both spin characters exist within the Cu film, in contrast to the expectations based on simple models.

The experiment has been performed on the TGM 1 beam line at BESSY. The experimental apparatus, which has been described elsewhere [10], was modified in order to orient a component of the light polarization along the surface normal. Spin-polarized spectra have been measured in magnetic remanence with a spherical energy analyzer couplied to a Mott spin detector. The measurements have been performed on a Cu/Co/Cu(100) layered system. The Cu(100) substrate was prepared *in*

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situ by sputtering and annealing cycles. A 20 monolayer thick fcc Co film has been grown on it at 100 °C. Cu ultrathin layers have been subsequently deposited on the Co film at room temperature. The base pressure of 10^{-10} mbar rose to 5×10^{-10} mbar during the evaporations. The characterization of Cu/Co/Cu(100) multilayer systems has already been extensively discussed in the literature [11,12]. Both fcc Co on Cu(100) and Cu on fcc Co(100) can be grown epitaxially. We could observe at all stages of the film growth a very good LEED pattern with a low background. The good quality of the overlayer growth is also demonstrated by the distinct and intense quantum well states. Ortega and co-workers [6,7] have pointed out that the presence of these states is perhaps the most critical test for the overlayer quality.

The evolution of the valence band states for increasing Cu coverage on Co(100) has been followed by measuring photoemission spectra at various photon energies. Spin-integrated photoemission spectra of the valence band region measured with 17 eV photon energy are presented in Fig. 1. The spectrum of clean Co shows the 3*d* emission near the Fermi level with a peak at 0.6 eV binding energy. With increasing coverage the Cu 3*d*-derived emission develops as an intense structure near 2.5 eV binding energy. Above about 5 monolayer thickness the Cu 3*d* spectral features essentially converge to those of the bulk at

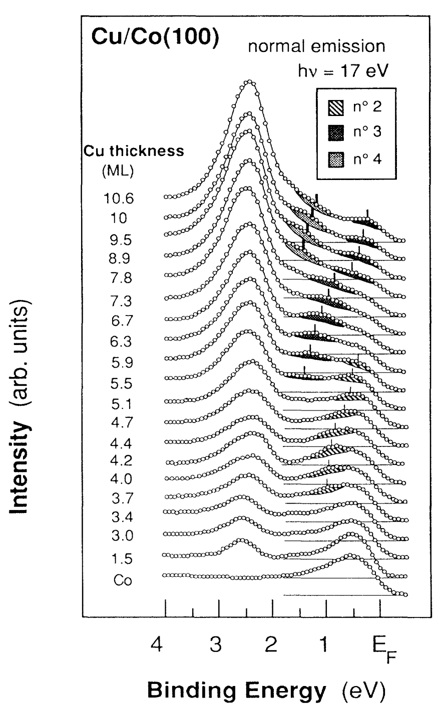

FIG. 1. Angle-resolved photoemission spectra of Cu overlayers on Co(100) for various thicknesses. The spectra have been measured for normal electron emission with 17 eV photon energy. The discrete overlayer states are labeled according to the definitions in Ref. [7].

all photon energies. The strongly delocalized states derived from the Cu *sp* levels are more sensitively affected by finite thickness of the overlayer up to higher coverage. The *sp*-derived states give rise to well-defined structures in the binding energy region within 2 eV from the Fermi level. These new states do not have a simple correspondence with the Cu(100) or the Co(100) bulk electronic structure. Their energy varies with the film thickness but they do not show any dispersion with change in photon energy. Their binding energies as well as the spectral shape are in agreement with the results of Ortega and co-workers [6,7] over the whole range of coverages. The normal emission spectra, as in Fig. 1, probe initial states with parallel wave vector component $k_\parallel=0$, i.e., corresponding to the $\bar{\Gamma}$ point. The observed structures have been explained taking into account the discretization of energy and perpendicular wave vector due to the finite size of the Cu film [6,7]. Within a framework of one-dimensional quantum well states a connection has been proposed between these states and the oscillatory magnetic coupling in Cu/Co(100) magnetic superlattice. Indeed the total wave vector of quantum well states can be expressed as $2\,(k_{\rm edge}-k_F)$, where $k_{\rm edge}$ is the wave vector of the nearest *sp* band edge (i.e., the X point) and k_F the Fermi wave vector. This is equivalent to the main RKKY vector along the [100] direction in Cu, arising from the "dogbone" structure of the Fermi surface [4]. Within a description based on the quantum well picture, the periodicity of the states observed near E_F can be related to the main oscillation period of the exchange coupling across Cu(100) layers that is 5–6 monolayers [12].

The main question to be addressed here is whether these states are spin polarized, in order to assess their role in the exchange coupling through Cu films. We have determined their spin polarization by spin-resolved photoemission. Some representative spin-resolved spectra are shown in Fig. 2. The exchange-split polarized states of the clean Co substrate are found within 3 eV from E_F. Little or no spin-dependent features are observed corresponding to the Cu 3*d* bands of the overlayer. In contrast, the Cu-induced structures in the region within 2 eV binding energy are found to be of spin-dependent character. Figure 2 shows in particular the development of these spin-polarized quantum well states between 6 and 9 monolayer coverage in some detail. In this range a pair of spin-down (minority) and a spin-up (majority) feature gradually shifts with increasing coverage towards the Fermi level. Spin-polarized states of the overlayer are observed in both spin channels up to a 10 monolayer thickness.

The occurrence and the polarization of quantum well states in Cu on Co(100) is not obvious according to simple considerations of bulk band structure. Ortega and Himpsel [6] predict that quantum well states near the Fermi level in Cu on Co(100) should be polarized but only of spin-up character, because Co has mostly spin-down states at the Fermi energy. However, a closer

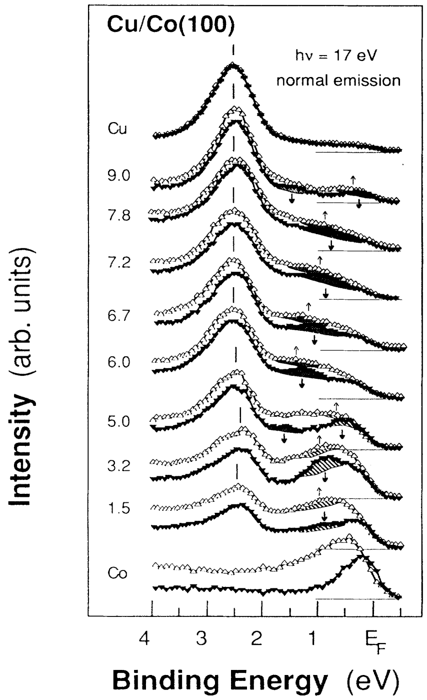

FIG. 2. Spin- and angle-resolved photoemission spectra of Cu overlayers on Co(100). The spectra are measured for normal emission with 17 eV photon energy. Open and closed triangles correspond to spin-up (majority) and spin-down (minority) electrons, respectively.

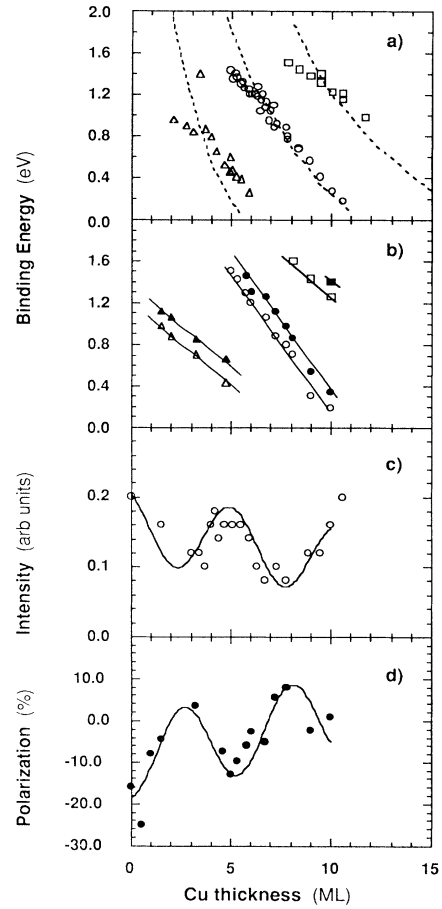

FIG. 3. (a) Binding energy of the features observed in the spin-integrated spectra (as in Fig. 1) as a function of the Cu overlayer thickness. (b) Binding energy of the features observed in the spin-resolved spectra (as in Fig. 2) as a function of the Cu overlayer thickness. (c) Spectral intensity at the Fermi level, after subtraction of the substrate contribution vs film thickness. (d) Spin polarization near the Femi level vs film thickness.

analysis of the Co band structure shows that no confinement in a strict sense should be expected in Cu at $\bar{\Gamma}$ in both spin directions near E_F and over large portions of the binding energy. Indeed the Co Δ_1 bands of both spin characteristics are strongly dispersive and cross the Fermi level along the Γ-X symmetry direction. True one-dimensional quantum well states can occur only in correspondence to the *s*-*d* hybridization gap, between 1 and 4 eV binding energy in the spin-down direction and between 2.5 and 5.5 eV in the spin-up direction [13]. This property of the Co band structure is consistent with the sharp and most intense spin-down Cu peaks which are experimentally observed below 1 eV binding energy. On the other hand, the observation of quantum well states of both spin characters over a binding energy region which extends beyond the gaps of the Co band structure suggests that the wave function localization and its energy dependence will have to be considered theoretically in a more detailed way. As proposed earlier [6,7], the interface potential may effectively act to partially confine within the film some *sp*-derived Cu states, even in the presence of some coupling to the degenerate levels of the substrate.

In the top panel of Fig. 3 the binding energies of the spin-integrated spectral features are compared with the calculations for one-dimensional quantum well states [7]. This theoretical model describes the development of the quantum well states with film thickness as derived from the bulk band edge wave functions. The model relies on assumptions which are valid in the asymptotic high-coverage limit and, as the results suggest, it might not be accurate in the low-coverage range. It provides, however, an overall acceptable description of the observed experimental trends. Figure 3(b) shows the energy position of the corresponding spin-polarized structure. States of opposite spin present an energy dispersion with coverage which is obviously similar to the one theoretically predicted for quantum well states, but the spin-up states are found at a slightly higher binding energy ($\Delta_E = 0.20 \pm 0.05$ eV) than their spin-down counterpart. Their energy separation is obviously a manifestation of the spin-dependent potential at the interface with the magnetic substrate. In a quantum well state model [6,7] it would be formally represented by a spin-dependent phase shift for reflection at the interface.

The results show that the discrete *sp*-derived states in the Cu overlayer become effectively polarized by the interaction with the magnetic substrate and thus are relevant for the long range coupling phenomena. By varying the film thicknesses, polarized quantum well states at a given energy periodically appear and disappear. By focusing attention to the region near the Fermi level one can observe a modulation of the intensity [Fig. 3(c)] and of the spin polarization [Fig. 3(d)]. Both curves have a similar periodicity which reflects their related origin. The maxima of the intensity curve occur when the spin-polarized peaks are close to E_F. In fact the spin-down features, which are sharper and more intense than the spin-up ones, account for most of the quantum well state intensity. Analogously, the minima of the polarization curve correspond to the spin-down states in proximity of the Fermi level. Conversely maxima appear in the polarization curve when the spin-up states are closer to E_F. The wave vector of the oscillations of both curves, about 5–6 monolayers, corresponds to the main RKKY wave vector along the ⟨100⟩ direction associated with the "dogbone" Fermi surface.

The existence of discrete and polarized states in both spin channels indicates that a distribution of the moments, in real space, within a Cu film of a given thickness is due to the interplay between the differently modulated charge density of both spins. The induced polarization within the Cu film might be qualitatively analogous to the oscillatory profile calculated for the hyperfine field by KKR Green's function method in multilayers [14], although the symmetry of the overlayer system is reduced by two inequivalent boundary conditions. The magnetic moment resulting from the polarization of the *sp* states is expected to be extremely small, of the order of $0.1\mu_B$ or less. Moreover the oscillating sign of the spin density further reduces the total moment if averaged over the whole film. The induced polarization effect on the nonmagnetic overlayer is therefore subtle and essentially restricted to a few specific electronic states. This can explain why other spin-polarized measurements which do not discriminate on the electron energy and symmetry [9] have not been able to detect the long range oscillatory behavior of the polarization in nonmagnetic overlayers.

In summary, we have detected spin-polarized electronic states in Cu films on a magnetic Co substrate. These states have been observed up to 10 atomic layers thickness. The photoemission intensity and polarization at the Fermi level determined by these spin-dependent states display a periodicity with the film thickness correlated with the oscillations of the magnetic coupling.

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